Temperature dependence of the magnetic excitations in ordered and disordered $Fe_{72}Pt_{28}$

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We have performed inelastic neutron scattering measurements of the long-wavelength spin wave excitations of both ordered and disordered $Fe_{72}Pt_{28}$ single crystals below their critical temperatures, $T_C=510$ and 375 K, respectively. The spin waves followed the expected $E=Dq^2$ dependence, and the temperature-dependent spin stiffness D decreased as $(T/T_C)^{5/2}$, as expected for an isotropic ferromagnet. The extrapolated zero-temperature spin stiffness was $D=98(4)$ meV Å^2 and 107(1) meV Å^2 for the disordered and ordered alloy, respectively. These values are significantly higher than the zero-temperature stiffness as determined by magnetization measurements.

INTRODUCTION

An understanding of the Invar effect has been suggested via recent band-structure calculations and experiments, which indicate that the magnetic moment of certain $3d$ alloys is unstable with respect to a change in volume.[1] The Invar effect originally referred to a magnetically induced suppression of the thermal contraction, first observed in disordered $Fe_{65}Ni_{35}$. Fe_3Pt alloys also display the Invar thermal expansion anomaly, with the added advantages that Fe is the sole magnetic component in the alloy, and that the Invar effect occurs in both atomically ordered and disordered Fe_3Pt (hereafter referred to as o-Fe_3Pt and d-Fe_3Pt, respectively). The thermal expansion anomaly associated with the magnetic ordering is much stronger than in $Fe_{65}Ni_{35}$, particularly for d-$Fe_{72}Pt_{28}$, where the thermal expansion coefficient changes from 1×10^{-5} K^{-1} at ≈40 K above T_C to -3.5×10^{-5} K^{-1} at ≈30 K below T_C.[2]

Another manifestation of the Invar effect is the discrepancy that exists between the magnetic excitation spectrum determined directly by neutron scattering and that inferred from magnetization measurements. For isotropic ferromagnets,[3] of which these cubic alloys are good examples, the dispersion relation is quadratic in the small-q regime, $E=Dq^2$, where D is the spin wave stiffness. The leading order temperature dependence of the magnetization is

$$M(T)=M(0)(1-BT^{3/2}), \tag{1}$$

where in conventional spin wave theory

$$B=\frac{\zeta(3/2)g\mu_B}{M(0)}\left(\frac{k_B}{4\pi D}\right)^{3/2}. \tag{2}$$

The magnetization at $T=0$ is $M(0)$; k_B is the Boltzmann constant; and ζ is the Riemann zeta function [$\zeta(3/2)\simeq2.612$]. The dynamic interaction between spin waves, which arises from the fact that multiple spin waves are not eigenfunctions of the Heisenberg Hamiltonian, decreases the spin wave energies and lifetimes. As a result, the temperature dependence of the spin stiffness is predicted to decrease to leading order as $T^{5/2}$,

$$D(T)=D(0)(1-AT^{5/2}). \tag{3}$$

It is thus possible to relate the spin wave stiffness to bulk magnetization measurements [using Eq. (1)]. In all Invar systems, there appears to be a substantial discrepancy between the spin wave stiffness as determined by bulk magnetization and by neutron scattering. In particular, the magnetization decreases more rapidly with increasing temperature than would be predicted by counting magnons as observed by neutron scattering. We have been studying a number of Invar and non-Invar systems in an effort to resolve this discrepancy.[4,5]

Previous inelastic neutron scattering work on o-$Fe_{72}Pt_{28}$ (Ref. 6) showed a 20% discrepancy between the decrease in magnetization calculated from applying spin-wave theory to the neutron data and the experimentally determined bulk magnetization. In this paper, we present the results of unpolarized inelastic neutron scattering measurements of the magnetic excitations in ordered and disordered $Fe_{72}Pt_{28}$. We find that there is indeed a significant difference between the spin wave stiffness as determined by bulk techniques and neutron scattering, for both the ordered and disordered alloy.

EXPERIMENT

The scattering measurements were performed at the BT-2 triple-axis spectrometer of the NIST Research Reactor with $21'-10'-10'-20'$ collimation before and after the monochromator and analyzer, respectively, and a pyrolytic graphite (PG) filter before the monochromator to remove higher-order wavelength contamination. The incident neutron energy was fixed at 13.7 meV (2.444 Å). Both the

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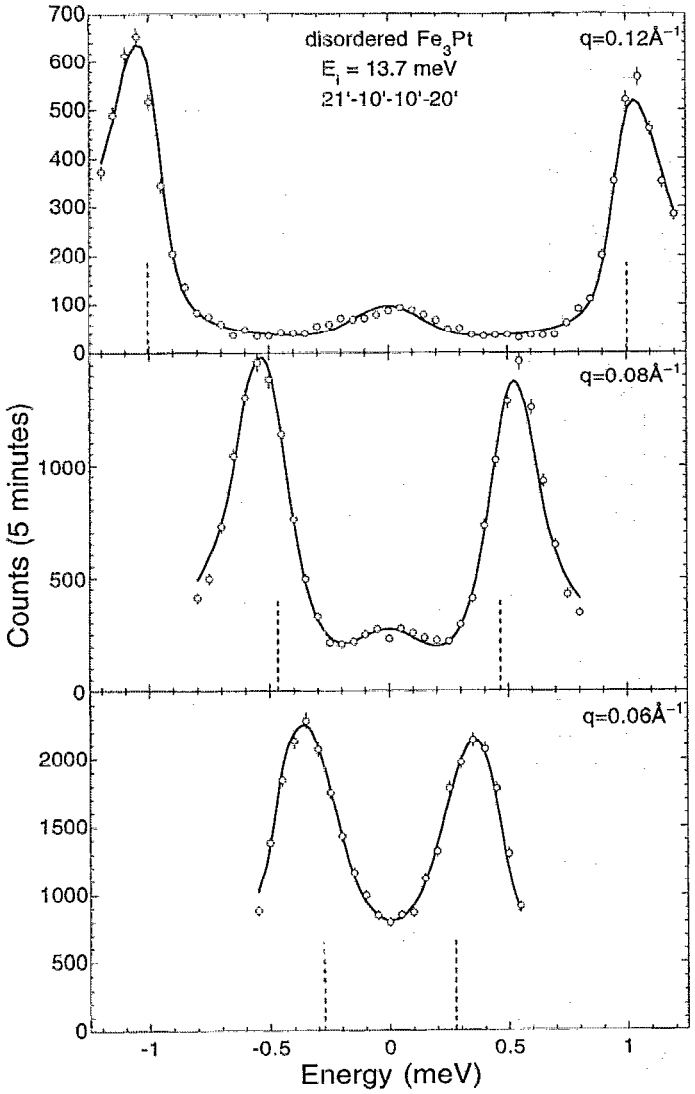

FIG. 1. Constant-q scans for disordered $Fe_{72}Pt_{28}$ about 000 at room temperature with momentum transfer $q=0.12$ Å^{-1} (top), 0.08 Å^{-1} (middle), and 0.06 Å^{-1} (bottom). The solid lines are best fits to the data of Lorentzian-broadened magnons and an elastic central peak convolved with the instrumental resolution function. The scans are limited in energy by the kinematic constraints on scattering about 000.

monochromator and the analyzer used the PG 002 reflection. In this configuration, the energy resolution is 0.33 meV, full width at half maximum (FWHM), the longitudinal q resolution in the scattering plane is ≈ 0.01 Å^{-1} FWHM, and the vertical resolution is ≈ 0.22 Å^{-1} FWHM.

The samples were approximately 20-g single crystals of $Fe_{72}Pt_{28}$. Atomically ordered and disordered $Fe_{72}Pt_{28}$ have Curie temperatures of $T_C=510$ and 375 K, respectively. The temperature was varied between 450 and 100 K using a closed-cycle He refrigerator. All the measurements were made well above the premartensitic transition that occurs in $Fe_{72}Pt_{28}$ below $T\approx 70$ K.

We measured the energy dependence of the scattering cross-section at momentum transfers in the range $0.05 \leq q \leq 0.17$ Å^{-1}. The inelastic scans were made about the 000 position, which gives several advantages to the measurements. First, the magnetic excitations can be observed without a significant contribution from lattice excitations. Second, we have available the greatest possible intensity for the excitations around 000, where the magnetic form factor is unity. At the 111 reciprocal lattice point, for instance, the magnetic form factor for Fe is 0.4,[7] and thus the scattering intensity will be $\approx 16\%$ of that observed about 000. Finally, we will be able to make a direct comparison of these results to planned polarized beam measurements about 000, which can be made on these samples only around 000 due to constraints on the orientation of the sample in the required applied magnetic field.[5] Nevertheless, there are several disadvantages to performing measurements about 000. First, the maximum achievable energy transfer increases linearly with momentum transfer, $E_{\pm}\approx \pm 2k_i q$, whereas the spin wave energy increases quadratically with q. Thus, for a particular spin stiffness, we are limited in the range of momentum transfer for which we may observe excitations (see Fig. 1). Second, the requisite small scattering angles necessitate tight angular resolution, and with the combined constraint of a restrictive energy range we also require good energy resolution, thus reducing the signal considerably.

RESULTS AND DISCUSSION

At each temperature, we measured the energy-dependent scattering cross-section for several momentum transfers. Typical data, shown in Fig. 1 for the disordered sample, reveal several characteristic features. The two peaks at finite neutron energy gain and loss are the magnon annihilation and creation peaks. The central elastic peak is due mainly to magnetic disorder scattering. As discussed above, data cannot be taken over the full extent of the magnon peaks due to kinematic constraints. The data were fit to a double-

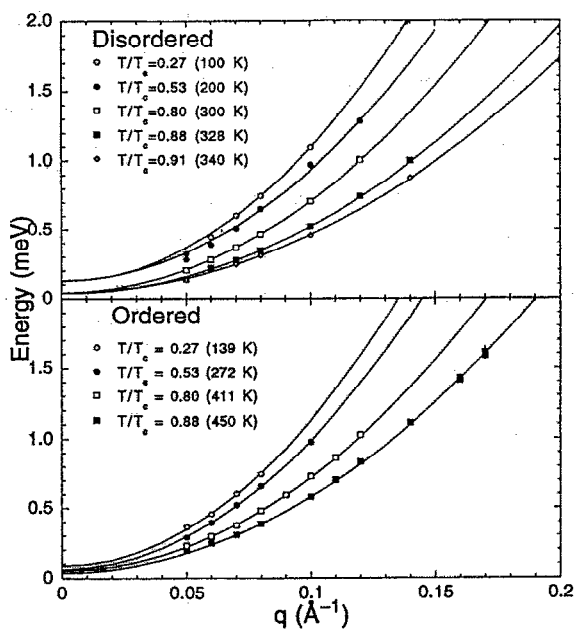

FIG. 2. Measured dispersion relations for magnons around 000 in disordered (top) and ordered (bottom) $Fe_{72}Pt_{28}$. The solid lines are the best fit of the data to a quadratic dispersion with a small gap, $E(q)=Dq^2+E_0$.

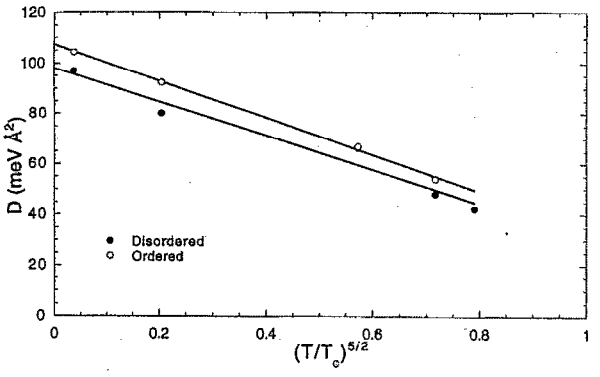

FIG. 3. The spin wave stiffness as a function of $(T/T_C)^{5/2}$ for disordered (●) and ordered (○) $Fe_{72}Pt_{28}$. The solid lines are linear best fits to the data; the slopes are the same, within experimental error, for both the ordered and disordered alloy.

Lorentzian spectral-weight function and an elastic central peak, which were numerically convolved with the triple-axis spectrometer resolution function.[8] For the purposes of the convolution, since the q-resolution is finite, the spin wave dispersion was assumed to be quadratic in q and the broadening was assumed to increase as q^4, according to spin wave theory; fits attempted with a quadratic dependence of the broadening on q were of poorer quality. In the data shown in Fig. 1, we obtained a spin stiffness $D(300\ \mathrm{K})=66.3(2)$ meV Å^2 and a line broadening $\Gamma(300\ \mathrm{K}, 0.08\ \text{Å}^{-1})=0.04$ meV HWHM. This intrinsic width is considerably smaller than the instrumental resolution.

From the fits to the measured scattering cross-sections, we have plotted the excitation energy versus momentum transfer for each temperature (see Fig. 2). The measured dispersions were then fit to a quadratic dispersion with a pseudo-gap that is due to dipolar interactions, $E(q,T)=D(T)q^2+E_0$. Obviously, from the quality of the fits, our assumptions in the fitting procedure of a quadratic dispersion are well founded. The temperature dependence of the spin-wave stiffness is shown in Fig. 3, plotted against $(T/T_C)^{5/2}$. The decrease in stiffness is consistent with a linear behavior over the range $0.27<T/T_C<0.91$, as indicated by the fit. The extrapolated zero-temperature spin stiffnesses are 98(4) meVÅ^2 and 107(1) meVÅ^2 for d-$Fe_{72}Pt_{28}$ and o-$Fe_{72}Pt_{28}$, respectively. The reduced temperature dependence of the spin stiffness scales with $D(0)$ for both alloys; the slopes of the fits in Fig. 3 are the same within experimental error. Thus, while the disorder causes a decrease in the magnetic ordering temperature, the spin dynamics of the alloys appears to follow the same Heisenberg-like behavior. The present $D(T)$ values for o-$Fe_{72}Pt_{28}$, as determined by neutron scattering, are in agreement with the room temperature neutron scattering spin stiffness [$D(300\ \mathrm{K})=85$ meVÅ^2][9] reported previously for o-$Fe_{72.7}Pt_{27.3}$. No previous results have been reported for the disordered system.

The zero-temperature spin stiffnesses for both the ordered and disordered alloys are significantly higher than the corresponding values determined from magnetization measurements, $D_m^{\mathrm{disord}}(0)=74$ meVÅ^2 and $D_m^{\mathrm{ord}}(0)=89$ meV Å^2. These values are about 20% less than the spin wave determinations, which is about the same relative difference as reported in previous measurements[6] on o-$Fe_{72}Pt_{28}$. The 20% discrepancy between the inelastic neutron and bulk magnetization spin stiffnesses is significantly less than the factor of two discrepancy that is observed for crystalline $Fe_{65}Ni_{35}$,[6] as well as other Invar systems such as amorphous $Fe_{86}B_{14}$,[3] despite the fact that the thermal expansion anomaly is much stronger in $Fe_{72}Pt_{28}$. Further work with polarized neutrons may allow us to characterize the nature of the excitations in these systems.[5]

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